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Resolving archaeological populations with Sr-isotope mixing models

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Abstract

Sr isotope analysis of tooth enamel is a useful provenancing technique to investigate the childhood origins and residential mobility of ancient people. However, where different geographical target regions have similar biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ it is often difficult to resolve the $^{87}\text{Sr}/^{86}\text{Sr}$ ranges of two different groups of people and establish what constitutes the local range at each site. Here we present a multi-period study from the Outer Hebrides, Scotland and an investigation of Neolithic and Early Bronze Age populations from the Yorkshire Wolds, NE England. The aim is to demonstrate that, despite complex human dietary strategies, simple mixing systems with only two end-members do occur in archaeological human populations in certain geological provinces and, despite overlapping $^{87}\text{Sr}/^{86}\text{Sr}$ ranges, it is possible to separate two populations based on the *structure* within the data set.

Keywords

Sr-isotopes; teeth; enamel; archaeology; Britain; migration; provenance; mixing diagrams.

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Introduction

Sr isotope analysis of tooth enamel is a useful provenancing technique to investigate the childhood origins and residential mobility of ancient people (e.g. Bentley and Knipper, 2005; Evans and Tatham, 2004; Ezzo and Price, 2002; Montgomery *et al.*, 2000, 2003; Price *et al.*, 2002, 2004). Enamel is a highly mineralised, acellular, biogenic apatite that is particularly resistant to post-mortem contamination and thus preserves the integrity of lifetime signatures (Budd *et al.*, 2000; Hoppe *et al.*, 2003; Montgomery, 2002; Trickett *et al.*, 2003). The $^{87}\text{Sr}/^{86}\text{Sr}$ value of enamel is derived from food and water ingested when the tissue was mineralising, that is, in early childhood. Food and water have $^{87}\text{Sr}/^{86}\text{Sr}$ values that reflect their geographical origin because Sr weathers from the host rock into groundwater, river water and overlying soils and is ultimately transferred unfractionated into plants and animals (Åberg, 1995; Capo *et al.*, 1998; Ericson, 1985; Price *et al.*, 2002). This provides the mechanism to establish whether a person or animal has an isotope ratio consistent with locally grown food or locally sourced water, and if this is not the case, to conclude that they either imported substantial resources from elsewhere (perhaps more likely in modern populations) or that they had moved from a different geological province at some time after the tooth had mineralised.

An ideal outcome is that the Sr isotope ratios of the population under study divide neatly into two (or more) visibly separate groups on a isotope plot. However, this rarely happens and investigators are frequently faced with a smear of data points with no discernible break and, perhaps, one or two obvious outliers (see for example Montgomery *et al.*, 2005; Schweissing and Grupe, 2000). In such a situation it is often very difficult to ascertain where one should draw a line between local people and

those from elsewhere or if only the outliers can confidently be ascribed non-local status. It is clearly more difficult to identify two populations if some degree of overlap, arising perhaps through similar biosphere values being available in their respective home regions, is present in the range of $^{87}\text{Sr}/^{86}\text{Sr}$ values (Price *et al.*, 2002). Given this potential for overlapping $^{87}\text{Sr}/^{86}\text{Sr}$ ranges in populations originating in different geographical regions, it is perhaps not surprising that it has often proved difficult in previous UK studies to resolve the respective ranges of two supposedly different populations (Evans and Tatham, 2004; Montgomery *et al.*, 2005). Several authors have advocated identifying local populations by the application of a statistical or numerical cut-off based on two or more standard deviations from the mean of bone or animal values (Bentley *et al.*, 2004; Grupe *et al.*, 1997; Horn and Müller-Sohnius, 1999). Whilst this may work well in the specific time period, region and site they are investigating, such a parametric method presupposes that the local population will produce a Gaussian distribution of $^{87}\text{Sr}/^{86}\text{Sr}$ values which, as in the case of lead isotope fields, is more likely to be the exception rather than the rule, rendering many statistical methods inappropriate (Baxter, 1999; Scaife *et al.*, 1996). Moreover, the number and type of end-members and degree of variation a population exhibits may change considerably as we move from different geological, geographical and even cultural milieu. The absolute value and the magnitude of the required cut-off will be very specific to the local area and its geology, rendering each site different.

Our current model for Sr isotope uptake from the geosphere in a maritime environment such as the British Isles, is that a community living on and sourcing their food from a single, homogeneous rock unit will nevertheless exhibit a range of $^{87}\text{Sr}/^{86}\text{Sr}$ values that fall between two end-members: the local rock and rainwater

(Evans and Tatham, 2004; Montgomery, 2002; Montgomery *et al.*, 2005). This may sound unrealistic and oversimplified, but it does not require that *types* of foods are restricted, only that they are all grown and grazed on just one type of bedrock plus rainwater. Rainwater has seawater of $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7092$ as its source and it can be ingested in variable quantities through food and drink. The $^{87}\text{Sr}/^{86}\text{Sr}$ value of rainwater in coastal regions is very similar to seawater but over landmasses, the incorporation of terrestrial dust aerosols will progressively alter the original marine signature (Capo *et al.*, 1998; Faure, 1986). The resulting change in $^{87}\text{Sr}/^{86}\text{Sr}$ will depend on distance from the coast and the $^{87}\text{Sr}/^{86}\text{Sr}$ value and Sr concentration in the dust sources and may vary seasonally depending on weather patterns (Åberg, 1995; Andersson *et al.*, 1990; Bacon *et al.*, 1995; Land *et al.*, 2000; Miller *et al.*, 1993). For example, Negrel *et al.* (2001) found that rainwater varied between 0.70901 and 0.71060 in the Massif Central region of SE France but had a mean value of 0.70943 weighted by % of rainfall.

Consequently, a sedentary, self-sufficient population will not define a normally distributed range of $^{87}\text{Sr}/^{86}\text{Sr}$ values *around* the local rock value but a range offset above or below the rock value (Montgomery *et al.*, 2005). The magnitude of the range will depend on how different the local rock Sr is from 0.7092: *i.e.* a population from a region of 0.715 rocks will, through personal choice, cultural practice and resource availability, have the opportunity to obtain an enamel $^{87}\text{Sr}/^{86}\text{Sr}$ value between 0.7092 and 0.715, that is, a weighted average of all inputs. Alternatively, a population subsisting on Cretaceous Chalk ($^{87}\text{Sr}/^{86}\text{Sr} \sim 0.707$) will only have the opportunity to fall within the range 0.707 – 0.7092 and will, therefore, define a much smaller spread of values.

The model becomes increasingly problematical in regions where several different rock types crop out locally or heterogeneous silicate rocks such as granites and sandstones occur. In silicates, Sr released through weathering may not be representative of the whole-rock value but will be dominated by unradiogenic Sr derived from the more easily weathered minerals such as calcite and feldspar (Åberg, 1995; Bau *et al.*, 2004; Blum *et al.*, 1993; Evans and Tatham, 2004; Jacobson and Blum, 2000). Drinking water sources (such as rivers, deep aquifers and springs) that are not derived directly from rain may also have $^{87}\text{Sr}/^{86}\text{Sr}$ values quite different from seawater if Sr has been dissolved from the host rocks (Montgomery *et al.*, 2006). In addition, through choice or necessity, populations may vary considerably in their food procurement strategies, level of sedentism and cultural practices and this suggests we are unlikely to find populations who have a predictable Sr ratio derived from only two dominant sources.

However, two of our recent case studies suggest that simple mixing systems with only two end-members *do* occur in archaeological populations and, despite overlapping Sr isotope *ranges*, it is possible to separate two populations based on the *structure* within the data set. We present the results of two case studies from very different geological provinces, which one would hypothesise, produce very different Sr isotope ratios in their respective indigenous populations. The first is a study of Neolithic and Early Bronze Age populations from the Yorkshire Wolds, NE England and the second from the Outer Hebrides (or Western Isles) of Scotland from the Neolithic to the Viking colonisation of the islands in the 8th – 9th centuries AD (Montgomery, 2002; Montgomery *et al.*, 2003) (Figure 1). What the structural differences found in the data

sets at both locations might tell us about the people, their geographical origins and food procurement strategies at each location will be discussed. Moreover, given the evidence in the literature that skeletal Sr concentrations do not reflect in any linear way the amount of Sr *ingested* (Burton and Wright, 1995; Elias, 1980), we discuss why and how some archaeological populations may define a clear two-way mixing line and what underlying factors may be ultimately controlling an individual's Sr uptake.

Methods and materials

Enamel samples were of core enamel only. Once childhood tooth mineralization is complete, core enamel is resistant to subsequent isotopic or elemental changes either during an individual's lifetime or during burial, whereas dentine equilibrates with the burial environment (Bocherens *et al.*, 1994; Budd *et al.*, 2000; Glimcher *et al.*, 1990; Hoppe *et al.*, 2003; Montgomery, 2002). To remove soil-derived particulate on the tooth surface, all enamel surfaces were abraded to a depth >100 µm with acid-cleaned, tungsten carbide dental burs. All adhering dentine and enamel-dentine junction tissue were also removed entirely. Dentine samples were removed from the tooth crown and all surfaces abraded with tungsten carbide dental burs.

Six samples of soil and rock were obtained from: the Iron Age burial context at Galson; the Norse and Bronze Age burial contexts at Cnip; strata identified as sealed Iron Age and Norse cultivation soils, at Galson and Cnip respectively; and a sample of gneiss from the Cnip headland. The gneiss was washed in water (Millipore Alpha Q) and ground to a coarse powder in a tungsten carbide ball mill. Modern plant samples were obtained from machair (Quaternary shell-sands), blackland (peaty soils

found between the coastal machair and the interior peat) and hill-land areas on South Uist. Plants were washed in water (Millipore Alpha Q, <1 ppb total heavy metal content), dried and powdered in a freezer-mill.

All enamel, dentine, plant and soil samples were transferred in sealed containers to the class 100, HEPA-filtered laboratory at the NERC Isotope Geosciences Laboratory (NIGL), Keyworth, UK. Enamel chips were washed ultrasonically in water (Millipore Alpha Q, <1 ppb total heavy metal content) to remove adhering particulate. No chemical decontamination was carried out as, due to the high resistance of enamel to post-mortem contamination, changes in the bulk $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of enamel samples following the use of such procedures has been negligible (Horn *et al.* 1994; Trickett *et al.* 2003). Approximately 2 g of each soil and rock sample was leached overnight with water (Millipore Alpha Q) and a further 2 g leached overnight in dilute acid (10% vol. acetic). Samples were centrifuged and the leachate pipetted out and dried down. Circa 50 milligrams of plant powder was dissolved in a combination of Teflon-distilled 16M nitric acid and Romil Super Purity hydrogen peroxide. The laboratory procedure used ion exchange chromatography and Teflon-distilled reagents to isolate the Sr prior to instrumental analysis. The full method of preparation and analysis is reported in Montgomery (2002). Sr concentrations and compositions were obtained by thermal-ionisation mass spectrometry (TIMS) using a Finnigan Mat 262 multi-collector mass spectrometer. $^{87}\text{Sr}/^{86}\text{Sr}$ was normalized to a NBS 987 value of 0.710250. The Sr contribution from within-run laboratory blanks was ≤ 150 pg. External reproducibility was estimated at $\pm 0.004\%$ (2σ).

Results

The Yorkshire Wolds

The precise nature of the Neolithic-Bronze Age transition and when sedentary agriculturalism commenced in Britain remain major questions in British archaeology and the archaeological aim of this study was to investigate diet and mobility within and between Neolithic and Bronze Age populations in prehistoric East Yorkshire (Cooper, 2004).

Figure 2 displays enamel data obtained from 22 individuals (23 teeth) excavated in the 19th century by J.R. Mortimer (1905) from two Late Neolithic and six Early Bronze Age funerary barrows located on the Cretaceous Chalk of the Yorkshire Wolds (Figure 1). The area has virtually no drift deposits and one, intermittent, water source: the Gypsy Race. The Chalk was deposited during Late Cretaceous times and records $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between 0.7075 and 0.7078 (McArthur *et al.*, 2001). Direct analyses of Chalk, chalk-derived soils and water from chalk aquifers in England have provided $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from 0.7075 to 0.7077 (Evans *et al.*, 2006; Montgomery, 2002; Montgomery *et al.*, 2005, 2006). According to our simple two-component model, a community subsisting entirely on food sourced from a chalk substrate plus an input from rainwater should define a spread of ratios from ~0.7075 to ~0.7092.

Although the two groups of data show almost total overlap in both Sr concentration and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, making it difficult to separate them based only on *individual* values, the structure within the two *data sets* differs markedly. The Bronze Age individuals split into two groups, both of which form discrete linear arrays (labelled A and B) that appear to converge on a lower end-member ~0.7075. Such arrays occur

when the samples contain variable mixtures of just two sources of Sr which have different ratios and concentrations (Faure, 1986). In contrast, the Neolithic individuals form a diffuse cloud of data points with $^{87}\text{Sr}/^{86}\text{Sr}$ values ranging between 0.7079 and 0.7102. The lack of a direct linear relationship between the samples implies the presence of more than two end-members.

Tooth enamel contains Sr that was ingested over a specific and restricted period of time during childhood when the tooth analysed was mineralising. In human terms, therefore, the Bronze Age data is consistent with both groups procuring resources *from only two* sources of Sr and a different mix of these two Sr inputs has resulted in enamel samples containing various mixtures of the two sources. Such variability could arise through necessity with seasonal availability of resources, personal food preferences or, given the imprecision inherent in archaeological dating, gradual change over longer timescales. Amongst the Neolithic population, however, no such control over resources seems to be present and the distribution of the data is indicative of resources having been procured from *more than two* Sr sources. This suggests that the earlier Neolithic population were more eclectic and opportunistic in their exploitation of resources and foraged over a wider geographical area beyond the Wolds.

For the Bronze Age population Line A has a radiogenic upper end-member ≥ 0.7120 . To date, there is little evidence that such values can be obtained from the biosphere overlying the Permo-Triassic and Jurassic sedimentary deposits immediately to the north and west of the Chalk but some evidence that such values may be obtained from the Carboniferous Coal Measures which lie further to the west (Table 2; Evans and

Tatham, 2004; Montgomery, 2002; Montgomery *et al.*, 2005, 2006). For Line A and B, Chalk could provide the common lower end-member of $^{87}\text{Sr}/^{86}\text{Sr} \sim 0.7075$. However, Line A could equally have a lower end-member of ≤ 0.7092 , which is approximately the value of rain and seawater. Seawater Sr could provide the upper end-member for Line B through exploitation of marine or coastal resources. Alternatively, terrestrial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios > 0.7086 have been obtained from the plants, soils and mineral waters in regions of Permo-Triassic and Jurassic sedimentary rocks to the north and west (Table 2; Evans and Tatham, 2004; Montgomery, 2002; Montgomery *et al.*, 2005, 2006).

We suggest, therefore, that both Bronze Age groups conform to our simple two end-member model for sedentary agricultural communities, farming on one type of geology with a secondary input of drinking water: one group inhabiting the Wolds and the second group the higher land of the Pennine foothills some distance to the west. The end-member for the lower mixing line could simply be drinking water (suggesting a sedentary lifestyle on the Wolds), or seawater (suggesting utilisation of coastal resources), or as suggested for the Neolithic population, regions immediately adjacent to the Wolds. It must, however, be remembered that quite different sources of Sr (*e.g.* sedimentary rock and modern seawater) may have the same $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and that whilst geochemically these may be identical, archaeologically the difference may be important. Our results may, for example, indicate that Bronze Age communities were following a rigidly controlled system of food production where crops were grown in one place and livestock grazed in another or one of seasonal transhumance. Exploitation of coastal and marine resources may explain the 0.7092 end-member in both Bronze Age groups. Equally, it could be that both groups were

regularly visiting the Wolds for cultural reasons, the area is rich in ceremonial sites such as henges, and thus coming together there at certain times of year. In this regard, it should be noted that for one Bronze Age male adult, two teeth were sampled. The canine tooth (0-5 years of age) plots on Line A whilst the later forming third molar tooth (7-12 years of age) plots on Line B suggesting a move in later childhood from one resource-group to the other. We believe it is unlikely that the mixing lines are evidence for a single migration event to the Wolds from elsewhere as all of the individuals analysed would have had to make this move as children whilst the specific teeth we analysed were mineralising in order to incorporate different mixtures of the two sources.

The Outer Hebrides

The Outer Hebrides are the most northwesterly islands of the British Isles (Figure 1). The island chain is geologically homogeneous (Lewisian gneisses and granites) and has a clearly defined geographic boundary (the sea) separating it from places of different geology (and hence isotope ratios) and, we may venture, different subsistence strategies. Whether the sea represented a barrier of any real significance in the periods in question and whether any social or dietary difference existed between Outer Hebrideans and people inhabiting the Inner Hebrides (*e.g.* the Isle of Skye) or the Scottish mainland is debatable but it clearly presents more of a physical obstacle than, for example, trying to define the area of land utilised by a community and their animals living in central England (Evans and Tatham 2004). Quaternary shell sands (machair) overlie much of the Lewisian gneiss on the west coast of the Outer Hebrides. The machair plain extends for up to 2 km inland and throughout

much of the prehistory was the focus for settlement and agriculture. The island interiors are relatively tree-less and predominantly composed of bare rock, standing water and uncultivable blanket peat that supports very little terrestrial wildlife (Armit, 1996). To date, all biosphere samples analysed (*e.g.* burial and ancient cultivation soils, modern plants, archaeological herbivores) have produced $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between 0.7092 and 0.7104 although leaches of the gneiss rock provided ~ 0.715 (Table 2; Montgomery *et al.*, 2003). Hebridean redshanks have produced $^{87}\text{Sr}/^{86}\text{Sr}$ values from 0.7092 to 0.7100 (Bullman, 2003). This has led to the hypothesis that the biosphere Sr of this maritime island chain is dominated by $^{87}\text{Sr}/^{86}\text{Sr} = 0.7092$ of marine (*e.g.* rainwater, fish, marine mammals, sea-splash, seaweed as fertilizer, food and fodder) and machair (*e.g.* grazing and arable crops) origin with only a limited contribution from the gneiss (Montgomery *et al.*, 2003).

Figure 3 displays archaeological enamel data from 20 Hebridean humans and four herbivores. The herbivores form a cluster on the left of the plot and the humans separate into two groups; one individual (labelled E) fits into neither group but falls within the field of data obtained from Neolithic Chalk burials transferred from Figure 2. For ease of reference we have termed the two groups machair dwellers and silicate dwellers. Machair dwellers and herbivores are defined by high Sr concentrations and Sr ratios within the range of biosphere values obtained from the Hebrides (*i.e.* 0.7092 – 0.7104). In contrast, silicate dwellers define a linear array, which appears to arise from mixing between two highly disparate silicate end-members with no input from machair, marine or marine carbonate sources. To date, we have found no biosphere sources on the Outer Hebrides that could provide either the high (>0.7130) or low (<0.7078) end-members. Such an array reflects the compositional range and isotope

features seen in silicate rocks of the Inner Hebrides and west coast of Scotland (Fowler *et al.*, 2004) leading to the hypothesis that the members of the silicate group are immigrants to the Outer Hebrides (Montgomery and Evans, 2006). Clearly, this conclusion would have been difficult to make for individuals A and K, which lie just outside the defined machair field, if only $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were used. Two Bronze Age burials from the Inner Hebridean islands of Mull and Arran also fall on the silicate mixing line (Figure 3) and are thus consistent with such a hypothesis.

The machair dwellers define a slightly sloping field suggesting that their Sr is predominantly of marine or machair origin (0.7092) with only a very small contribution from the gneiss (~0.715). Most archaeological human enamel analysed to date from England has Sr concentrations in the range 30-100 ppm (Evans *et al.*, 2006; Evans and Tatham, 2004; Montgomery, 2002; Montgomery *et al.*, 2003, 2005), including those excavated from inland chalk and limestone sites, which suggests the high concentrations obtained from the machair group (150-400ppm) are not simply a result of subsisting on food grown on soils overlying marine carbonate rocks. Such high concentrations coupled with marine Sr ratios as seen in this island population may, instead, be characteristic of people who cling to the coast and combine exploitation of marine resources such as seaweed with small-scale farming of marginal lands (Kvamme *et al.* 2004). The herbivores, also, are dominated by marine Sr, although they have much higher enamel concentrations than the humans. This is not unexpected in vegetarians (Bocherens *et al.*, 1994; Tuross *et al.*, 1989). The Hebridean herbivores do not follow the same trajectory as the humans but they do not consume the same diet. Clearly the Sr concentration in the herbivore enamel is not directly relevant to human diets, because people do not eat teeth, and meat and milk

contain much less Sr than skeletal tissues (Burton *et al.*, 2003; Burton and Wright, 1995; Tsalev, 1984). Moreover, not all dietary components will contribute equally to body fluids due to complex synergisms and antagonisms with other dietary components such as calcium, fibre and protein, which ensure that there is no simple direct transfer of Sr from the diet to the skeleton (Elias, 1980; Elias *et al.*, 1982; Burton and Wright, 1995; Burton *et al.*, 2003). It is, therefore, possible that the herbivores are the immediate dietary upper end-member for the human population and a proxy for the gneiss, despite falling on the left-hand side of the plot in Figure 3. The data from modern plants (Table 2) shows that blackland and hill-land grazing can provide $^{87}\text{Sr}/^{86}\text{Sr}$ values at the upper end of the Outer Hebridean biosphere range (*i.e.* 0.7092 – 0.7104).

Diagenesis

An alternative explanation for the presence of mixing lines in these studies would be that they are produced by postmortem incorporation of Sr from the burial environment. The unexpectedly high enamel Sr concentrations found in the Hebridean machair dwellers are a particular cause for concern although they do not exceed normal modern concentrations which are known to be geographically variable but are typically 50-300ppm (Aufderheide, 1989; Brudevold and Söremark, 1967; Budd *et al.*, 1998; Chaudhri, 1995; Elias, 1980; Elliott and Grime, 1993; Hancock *et al.*, 1989; Lee *et al.*, 1999; Montgomery, 2002; Montgomery *et al.*, 1999; Underwood, 1977). Is it possible, for example that the two groups are simply contaminated and uncontaminated samples rather than immigrant and indigenous humans? However, such a conclusion would require that human and herbivore enamel were being acted upon differently in the burial environment despite all being buried in machair soil, and

require the presence of biosphere sources to provide the necessary end-members for the silicate mixing line. In the Yorkshire Wolds study, all individuals were excavated from barrows on the Chalk and it would be difficult to make a case that diagenesis was only affecting the Bronze Age burials in this manner, especially since they had spent less time in the ground than the Neolithic burials.

For some of the teeth we analysed, we also sampled dentine from the crown of the tooth. There is considerable overlap in the mineralisation periods of enamel and primary crown dentine and neither tissue subsequently remodels or re-grows (Boyde, 1989; Fincham and Simmer, 1997; Sasaki *et al.*, 1997; Veis, 1989; Wieser *et al.*, 1996). Consequently, in modern individuals we can expect the Sr concentration and isotope ratio of these two tissues to remain very similar and this does appear to be the case even in individuals who have subsequently been exposed to different sources of Sr (Montgomery, 2002; Tsalev, 1984). Similarly, archaeological individuals that have not been buried, such as the Neolithic skeleton HAUG-1 (Table 1) which was removed from a chambered tomb in which we presume very limited exposure to soils and fluids took place, also show this pattern. However, in teeth excavated from soil burials it is usual for dentine to contain more Sr than enamel and this increase is normally coupled with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios intermediate between the enamel value and that of the burial soil (Budd *et al.*, 2000; Montgomery, 2002; Trickett *et al.*, 2003). Figure 4 shows paired enamel and dentine values for three teeth excavated from three different barrows. Diagenetic vectors are shown representing two-component mixing between the biogenic composition of the enamel (used as a proxy for biogenic crown dentine) and the mobile Sr in the burial environment, which has produced a postmortem shift in the crown dentine composition. The vectors appear to converge

on $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7082$, suggesting that the diagenetic Sr in the burial environment does not have the composition of pure Chalk. This may reflect the recorded use of “foreign” clay in the construction of the barrows (Mortimer, 1905). It appears that the contaminated dentine samples are not on the same mixing trajectory as the enamel samples whose biogenic isotopic integrity is intact. This discrepancy is also apparent when dentine samples from the Hebrides are plotted with the enamel data. Figure 5 shows that dentine samples are moving away from the enamel compositions and the diagenetic vectors are converging on $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7100$ rather than the biosphere lower end-member indicated by the enamel samples of 0.7092. The distribution of the dentine samples is also quite different to that of the enamel samples as there are no enamel data points within the diagenetic triangle defined by the dentine samples, which we would perhaps expect to find if enamel samples were being contaminated with Sr from the same source. Enamel samples equilibrating with burial Sr would not move down the silicate mixing line and then across but would move directly towards the value for diagenetic Sr. An exception to this is the sample from a neonatal infant (labelled G), from which only a partially formed deciduous tooth crown could be sampled. This tooth crown contained no fully mineralised enamel and was dark brown and relatively soft and it is possible that it did not have the same resistance to diagenesis as the fully mineralised, hard, translucent, white tissue analysed from the remaining samples in this study.

Discussion

A linear mixing relationship between human enamel samples requires not only close control over the isotope ratio but also over the concentration and, whilst it may be

perfectly reasonable to assume that random mixing of two food types may produce an assortment of *diets* on such a linear array with two identifiable end-members, because there is no opportunity for Sr to be added or taken away, obtaining such a relationship from biological skeletal tissues is considerably more complex. Although Sr concentrations in skeletal tissue may be dose-dependent (Comar *et al.* 1956; Boivin *et al.* 1996), experimental studies have concluded that there is no straightforward, linear, relationship between the amount of Sr in any given diet and the amount of Sr in the resulting tissue (Elias 1980; Burton and Wright 1995). Biological organisms are not rocks; physiological processes can act upon the amount of Sr that is finally deposited in tooth enamel at all the stages in its journey from the stomach to its final deposition in enamel. Moreover, Sr is not evenly distributed across all tissues; in mammals, most resides within the skeleton and given bone formation and turnover rates is unlikely to be in constant equilibrium with the diet. Enamel concentrations may be increased or reduced relative to the concentration in any one food type and not all dietary components will contribute equally; indeed some may not contribute at all. Why, then, should we expect to find such a relationship between a human population and its diet?

What is remarkable, given the discrepancy we might consequently expect between the concentration in the diet of an individual and the resulting concentration in their enamel, is the extremely good fit of the human samples on the mixing lines presented here. From a geochemical perspective, this is highly unlikely to arise from coincidence. As Faure (1986) asserts “*The goodness of fit of the data points to a straight line is a test for the validity of the mixing hypothesis and of the assumption that neither the Sr concentrations nor the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were modified after mixing had*

occurred". He is not writing about biological organisms, but the individuals in our case studies do indeed appear to be strongly related in such a simple binary relationship that can be explained through access to food and water that derives from two, and only two, sources of strontium. However, in order for these individuals to lie on the mixing line, not only their isotope ratio but also the concentration of Sr in their enamel must be very closely controlled by the two geological sources providing the dietary Sr and *not modified subsequently*. Such a finding clearly has implications for studies that use Sr/Ca ratios as an indicator of trophic level and food sources because, as already suggested by Burton *et al.* (2003), it raises the possibility that in certain locations geological, rather than biological, processes will control the amount of Sr in skeletal tissue.

For the Yorkshire Wolds study, archaeological dating and funerary evidence accords well with the geochemical evidence; they independently separate the individuals into the same two period-based groups suggesting the difference in food procurement strategy is a change through time (Cooper, 2004). Despite the strong linear relationship between the members of the Hebridean silicate group, radiocarbon dates from these individuals span nearly the whole of the first millennium AD and there is little archaeological evidence to link them together. As previously mentioned, one possible region that might provide such mixing and migration opportunities is the Tertiary Volcanic Province (Figure 1) where young basaltic rocks have extruded through the ancient Lewisian crust, as on the nearby Isle of Skye and Scottish mainland (Fowler *et al.* 2004). Unfortunately, skeletal remains of comparable date available for analysis from such areas are either non-existent or extremely rare. Such a linear relationship may simply be a feature of this unique geological setting, as linear

arrays are certainly not routinely observed in archaeological studies (*e.g.* Schweissing and Grupe 2003). Moreover, such a large spread in isotope ratios within a single population is unusual and makes it very difficult to apply a statistical cut-off value to identify immigrants in such geological provinces. However, the majority of published studies have been unavoidably biased towards areas where bone survives, and these are far more likely to be regions of alkaline rock, such as chalk and limestone, than silicate rocks, which frequently produce acidic soils that are not conducive to good bone preservation. Alternatively, the factor linking these individuals may be that they originate from quite different geographical locations, but that all locations are in regions of silicate rock where there is no input from either marine carbonates or coastal marine Sr, such as the Scottish mainland. This study is still underway but irrespective of where these individuals originated from the data appear to provide evidence for an enduring migration stream to the Outer Hebrides.

Conclusions

$^{87}\text{Sr}/^{86}\text{Sr}$ mixing models provide a useful method of separating archaeological populations that have overlapping isotope ranges based on the structure within the data set. The method is based on the identification of dietary end-members using simple graphical means rather than using inferential statistical procedures to define populations by means of their $^{87}\text{Sr}/^{86}\text{Sr}$ ranges and standard deviations (Baxter *et al.*, 2000). Whilst such statistical methods may work well for the site where they are developed, it is clear from the data presented here that they may not be transferable to other studies in different geological settings because the underlying assumptions about dietary Sr might be incorrect and the data may not be normally distributed.

Whilst we acknowledge that the graphical method may not work in regions of complex, heterogeneous geology or for populations that have multiple sources of dietary Sr, the studies presented here show that in some regions the bedrock geology appears to exert a close control over both the Sr concentrations and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in human tooth enamel.

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Figures

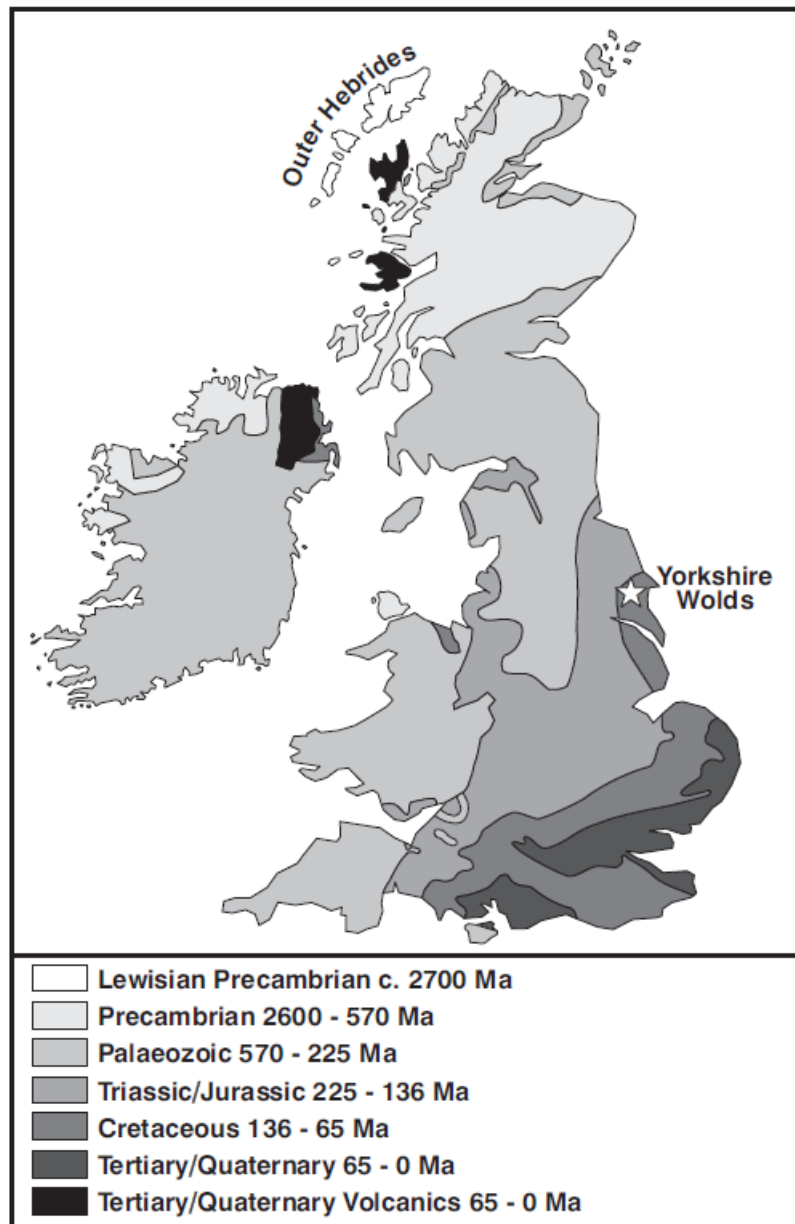


Figure 1. Simplified geology map of Great Britain and Ireland showing the location of the Outer Hebrides and the Yorkshire Wolds.

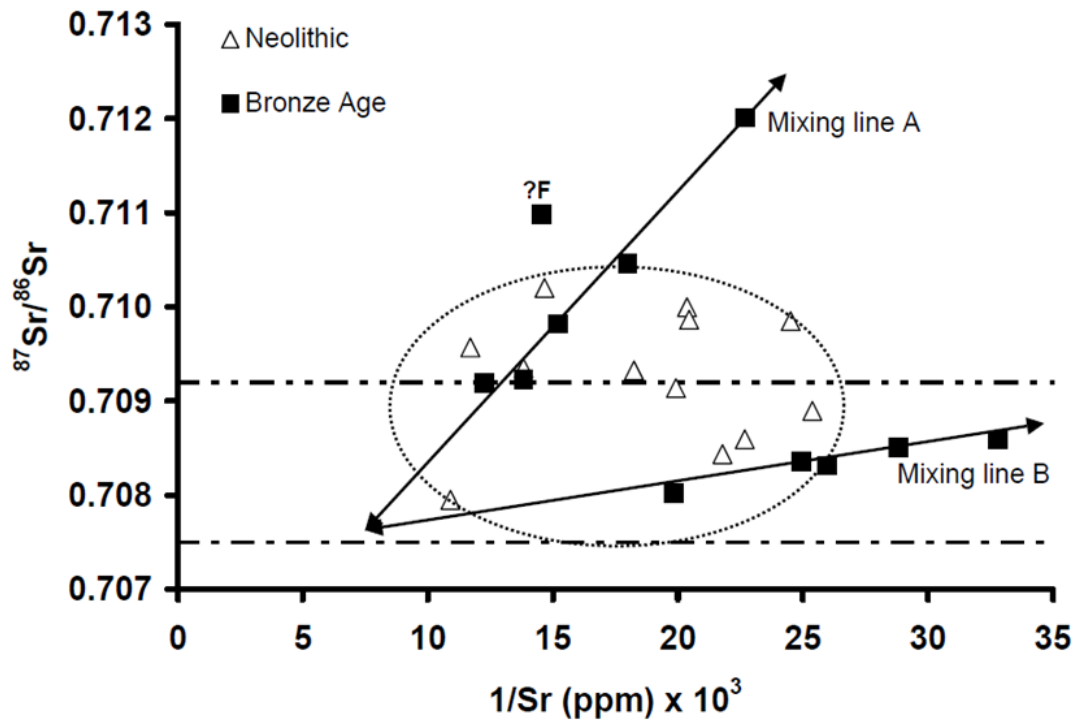


Figure 2. Human enamel data from Neolithic and Bronze Age barrows of the Yorkshire Wolds. The horizontal dotted lines indicate possible end-members: the upper line is seawater (~0.7092) and an approximation for rainwater; the lower line on which both mixing lines appear to converge is the value for English Cretaceous Chalk (Evans *et al.*, 2006; McArthur *et al.*, 2001; Montgomery, 2002; Montgomery *et al.*, 2005). For the upper mixing line $r^2 = 0.9789$, for the lower mixing line $r^2 = 0.9364$. The data point labelled ?F is the only skeleton plotted identified as being of possible female sex. 2σ errors are within symbol.

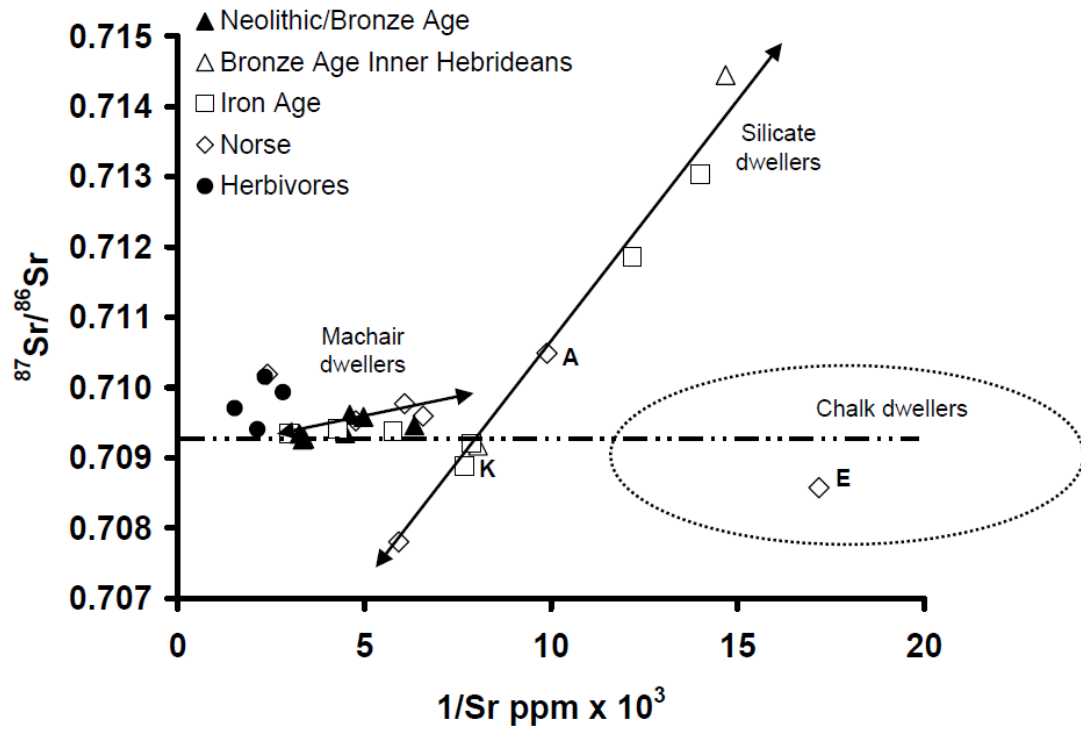


Figure 3. Human and herbivore enamel data from the Outer Hebrides, Scotland. The lower horizontal line denotes the seawater (~ 0.7092) end-member. For the silicate mixing line $r^2 = 0.9851$. The oval approximately replicates the field of Neolithic data from Figure X. All individuals were excavated from Outer Hebridean islands apart from the two Inner Hebrideans (Δ) from Arran and Mull. 2σ errors are within symbol. Data source: Table 1; Montgomery, 2002; Montgomery *et al.*, 2003; Parker Pearson *et al.*, 2005.

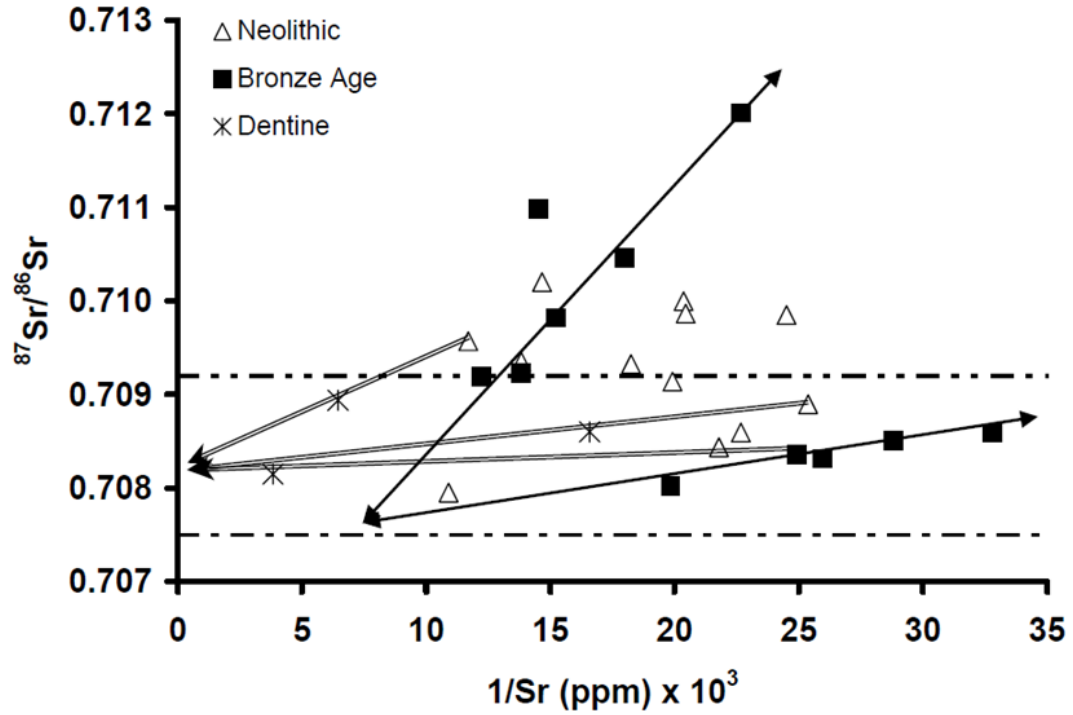
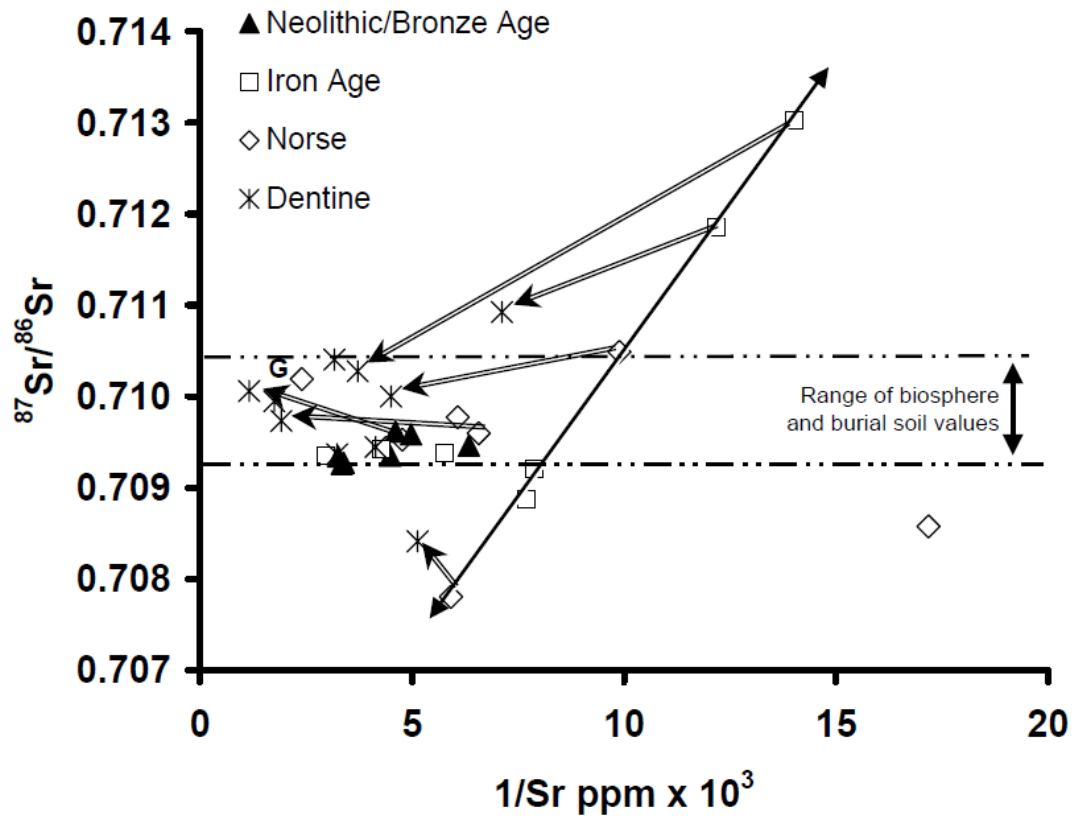


Figure 4. Human enamel and dentine data from Neolithic and Bronze Age barrows on the Yorkshire Wolds. Diagenetic vectors are shown between the lower concentration enamel and the higher concentration dentine value of three enamel-dentine pairs from three different barrows. They appear to converge on $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7082$ rather than the Chalk value of ≤ 0.7075 . 2σ errors are within symbol.



Tables

Table 1. Sr isotope ratio and concentration data for archaeological teeth. Teeth are identified by the following abbreviations: incisor (I); canine (C); premolar (P); molar (M); maxillary (^{1,2,3}); mandibular (_{1,2,3}); left (L); right (R); lower case letters indicate deciduous dentition.

Period	Site	Skeleton No.	Age	Sex	Tooth	Tissue	⁸⁷ Sr/ ⁸⁶ Sr	Sr ppm	1/Sr*1000
<i>Yorkshire Wolds</i>									
Neolithic	Towthorpe 273 (Duggleby Howe)	DH1/2/G	Adult	Male	I ² R	enamel	0.710201	68	14.66
		DH2/73/I	Adult	Male	M ² L	enamel	0.709570	86	11.70
						dentine	0.708938	155	6.45
		DH3/69/3	Adult	Male	P ₁ L	enamel	0.709138	50	19.92
		DH4/74/K	Adult	Male	P ₁ R	enamel	0.708594	44	22.68
		DH5/76/M	Adult	Male	P ₂ R	enamel	0.709997	49	20.37
		DH6/72/H	Juvenile	n/k	m ₂ R	enamel	0.709349	72	13.83
		DH7/75/L	Adult	Male	P ¹ R	enamel	0.709849	41	24.51
	Calais Wold 275	CW8/7	Adult	Male	P ² L	enamel	0.708896	39	25.38
						dentine	0.708600	60	16.58
		CW9/9	Adult	Male	M ² R	enamel	0.709865	49	20.45

Bronze Age	Calais Wold 100	CW10/8?	Adult	Male	C ¹ L	enamel	0.708437	46	21.79	
		CW11/3	Adult	Male	P ₁ R	enamel	0.709325	55	18.25	
		CW12/?	Adult	Male	P ₁ L	enamel	0.707952	92	10.91	
		Calais Wold 23	CW13/2	Adult	Male	P ₁ R	enamel	0.712010	44	22.68
		Calais Wold 23	CW14/1	Adult	Male	M ³ R	enamel	0.708023	50	19.84
		Towthorpe 73	TP15/1	Adult	Male	C ¹ R	enamel	0.709195	82	12.24
		Towthorpe 72	TP16/1	Adult	?Female	C ¹ L	enamel	0.709227	72	13.83
		Towthorpe 3	TP17/D	Adult	Male	P ² R	enamel	0.710458	56	17.99
		Aldro 116	AL18/1	Adult	Male	M ² R	enamel	0.710981	69	14.53
			AL19/4	Juvenile	n/k	C ¹ R	enamel	0.708315	39	25.97
							dentine	0.708149	261	3.83
			AL20/6	Adult	Male	P ₂ L	enamel	0.709818	66	15.22
			AL21/2	Adult	Male	P ¹ R	enamel	0.708512	35	28.82
			AL22/5	Juvenile 7-12yrs	n/k	M ₁ L	enamel	0.708588	31	32.79
			AL23/3	Adult	Male	P ₂ L	enamel	0.708361	40	24.94
<i>Hebrides</i>										
Neolithic	Haugabost, Harris	HAUG-1	Adult	n/k	M ₂ R	enamel	0.709340	307	3.26	
Bronze Age	Cnip, Lewis	BA-1	Adult	Male	C ¹ R	dentine	0.710403	316	3.17	
	Tormore, Arran	ET54	Adult	Female	M ¹ R	enamel	0.714447	68	14.68	
	Ardachy, Mull	MULL-1	Juvenile 15-20yrs	n/k	M ₁ L	enamel	0.709173	124	8.04	
						dentine	0.709815	555	1.80	

Iron Age	Kilpheder, S. Uist	SUK-1	Adult	Female	M ₂ L	enamel	0.708876	130	7.69
		SUK-2	Adult	Female	M ₁ L	enamel	0.709206	127	7.88
	Galson, Lewis	Gals-93	Adult	?Male	P ₁ R	dentine	0.710290	269	3.72
		Gals-96	Adult	?Male	P ₁ R	dentine	0.709380	309	3.23
		Gals-II	Adult	Female	P ₁ L	dentine	0.709459	242	4.14
		Gals-IV	Adult	Female	P ¹ L	dentine	0.710940	141	7.12
Norse	Cnip, Lewis	A	Adult	Female	P ¹ L	dentine	0.710014	222	4.50
		B	Juvenile ~6yrs	n/k	I ¹ L	dentine	0.709746	522	1.92
		C	Adult	Male	P ₁	dentine	0.710074	865	1.16
		D	Adult	Male	P ¹ R	dentine	0.708426	195	5.13
		F	Infant	n/k	di ¹ L	dentine	0.709961	572	1.75

Table 2. Sr isotope ratio and concentration data for modern plants. Sr concentrations should be regarded as estimates only as they are dependent on water content.

Location	Sample No.	Geology (bedrock or drift)	$^{87}\text{Sr}/^{86}\text{Sr}$	Sr ppm	Longitude	Latitude
South Uist	Frobost 309	Dry machair	0.709220	21	-7.4052	57.2034
	Frobost 309	Dry machair	0.709220	18	-7.4052	57.2034
	Milton 310	Machair	0.709219	56	-7.4006	57.2164
	Milton 310	Machair	0.710019	24	-7.4006	57.2164
	Milton 311	Low machair	0.709207	87	-7.4006	57.2164
	Milton 310a	Dry machair	0.709176	141	-7.4006	57.2164
	Milton 314	PreCambrian Gneiss - blackland	0.710289	62	-7.4006	57.2164
	Bornish 321	PreCambrian Gneiss - blackland	0.709449	45	-7.4015	57.2430
	Bornish 322	PreCambrian Gneiss - hill-land	0.710396	31	-7.4015	57.2430
Yorkshire	Hull-1	Cretaceous Chalk	0.708489	38	-0.4482	53.8417
	Hull-2	Permo-Triassic Sandstones	0.708997	24	-1.3128	53.7653
	Hull-3	Palaeozoic Magnesian Limestone	0.708637	55	-1.3453	53.8167
	Hull-4	Triassic Mudstones	0.710181	21	-0.8255	53.8298
	Hull-5	Permian Mudstones	0.709372	25	-1.1504	53.7696
	Hull-6	Jurassic Lower Lias	0.709691	30	-0.6593	53.2743
	Hull-7	Palaeozoic Carboniferous Coal Measures	0.714427	7	-1.8061	53.6961
	Hull-8	Palaeozoic Carboniferous Coal Measures	0.712636	8	-1.5525	53.6145
	Scar-1	Lower Jurassic clays and sands	0.709509	12	-0.6922	54.3386

Scar-2	Lower Jurassic clays and sands	0.709693	0.4	-0.6802	54.4255
Scar-3	Lower Jurassic clays and sands	0.708707	30	-0.8328	54.1178
Scar-5	Lower Jurassic clays and sands	0.709366	87	-0.9063	54.0783